

Fuel cleanup systems for fusion fuel processing

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The fuel cleanup unit (FCU) of a fusion fuel processing system receives gas from the torus evacuation system which is composed of hydrogen isotopes (deuterium and tritium) contaminated with a few to possibly around 20% of impurities (methane, helium, protium, water, nitrogen, carbon oxides, ammonia, etc.). Because the usual processing step subsequent to the FCU, the cryogenic isotope separation system (ISS), operates at about 25 K, the FCU must remove all impurities which freeze at this temperature to prevent ISS plugging. This includes all impurities other than helium and protium. This first function of the FCU is termed *purification*. The collected impurities contain a substantial quantity of tritium largely in the form of methane (and higher hydrocarbons), water and ammonia. This tritium must be recovered from the impurities before the remaining non-radioactive components may be discarded. This second function is referred to as *tritium recovery*. To accomplish these two functions a number of techniques have been proposed and investigated. This paper presents these techniques and identifies the merits and range of utility of each method.

1. Introduction

Fusion reactor exhaust, though composed of mostly hydrogen isotopes (Q_2), will contain a few to possibly around 20% of impurities. Examples of impurities are hydrocarbons (usually modelled as methane), ammonia, water, oxygen, argon, nitrogen, nitrogen oxides, carbon oxides and helium. All of these compounds, except helium, have higher boiling and freezing points than Q_2 . These impurities degrade plasma conditions

and, if introduced into a cryogenic isotope separation system (ISS), would freeze, resulting in plugs. Other streams which will require Q_2 purification include neutral beam, pellet injector and blanket tritium recovery gases.

It is the job of the fuel cleanup unit (FCU) to remove impurities upstream of the ISS. Moreover, some of the impurities, notably hydrocarbons such as methane, will have tritium substituted at sites normally occupied by hydrogen. This tritium cannot be dis-

Table 1
Examples of FCUs

Impurity removal process	Tritium recovery process	Ref.
Uranium carbides, nitrides, oxides	Occurs in impurity removal step	[6]
Cryogenic molecular sieve	Oxidation, water collection, water reduction (uranium)	[6–8]
Palladium permeator	Oxidation, water collection, water reduction (electrolysis)	[24,25]
Cryogenic molecular sieve	Oxidation, water collection, palladium membrane reactor	[26]
Cryogenic molecular sieve	Palladium membrane reactor	[26]
Cryogenic molecular sieve	High temperature catalytic exchange	[23]
Palladium permeator	Shift Catalyst/permeator	[12]

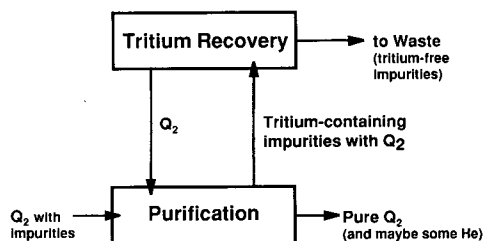


Fig. 1. FCU functionality.

carded, but must be recovered for later use. There are, then, two functions of the FCU, (1) to remove impurities from a stream of hydrogen isotopes, and (2) to recover the tritium bound to the impurities.

The function of an FCU can be accomplished in a variety of ways. Table 1 lists seven examples of complete FCU designs which have been proposed. In almost all cases the FCU operates in two stages. The first stage is *purification* which operates continuously (or semi-continuously), accepting gas, e.g., from the torus vacuum system and exhausting pure hydrogen isotopes to the ISS. The second stage is *tritium recovery* which extracts tritium from the impurities collected during purification and returns it to the fuel cycle for further use. Most tritium recovery schemes operate in a batch (or semi-batch) mode. The overall functionality of an FCU is shown schematically in fig. 1. In the following paragraphs methods for performing purification and tritium recovery are presented.

2. Purification schemes

In the following paragraphs four ways for removing impurities from a stream of Q_2 will be discussed, namely cryocondensation, hot metal beds (getters), cryogenic molecular sieve and palladium permeators.

2.1. Cryocondensation

This purification technique was first considered for the Tritium System Test Assembly (TSTA) at Los Alamos National Laboratory (LANL) in the late 1970s [1]. For this scheme four successively lower temperatures are used to collect impurities by freezing. The temperatures chosen were 180, 110, 45 and 25 K which were intended to collect water, ammonia, methane and everything else (except He), respectively. The result would be a stream of pure Q_2 and He. Expected operational difficulties and concerns about freezer

plugging and blowby resulted in the abandonment of this method.

Recently these principles have been applied in an innovative fashion and dubbed as the cryogenic diffusion pump by Hemmerich and Kussel [2]. Their investigation which is being conducted at the Joint European Torus (JET) Active Gas Handling System is directed toward development of a vacuum pumping system, but it may also serve to purify, as well as pump, the torus exhaust.

2.2. Hot Metal Beds

Hot metal beds were proposed for the TSTA FCU in the late 1970s. This method is unique in that both purification and tritium recovery are accomplished in one step. The Q_2 and impurities are passed over hot uranium (1170 K) which reacts to form solid carbide, nitride and oxide, leaving the original Q_2 and the Q_2 from the impurities in the gas phase. Another purification technique must be used with hot metal beds to collect unreactive impurities such as argon.

Though this system is attractive in its simplicity of operation, it has major drawbacks. These are: (1) it is difficult to reliably achieve and maintain 1170 K in a vacuum jacketed vessel, (2) the permeation rate of Q_2 through stainless steel is substantial at 1170 K, and (3) large amounts of radioactive waste would be produced since the uranium is used irreversibly. Schira and Hutter [3] have shown that even if all these problems are solved, it is difficult to remove impurities to below 1 ppm in a hot uranium bed. For these reasons this option is not viewed as being viable for large fuel processing requirements such as those expected for machines such as the International Thermonuclear Experimental Reactor (ITER). Recent work in this area by Ache et al. [4], however, may prove that this is a viable alternative for small processing flowrates where radioactive waste is not a major concern.

Albrecht, et al. [5,27] have reported on materials other than uranium that have also been investigated for performing hot metal bed purification such as SAES and HWT getters.

2.3. Cryogenic molecular sieve beds

This is the currently installed and operating method for purification at TSTA [6]. The Q_2 and impurities are passed through 77 K molecular sieve (Linde 5A). All impurities other than helium are adsorbed on the sieve, leaving only Q_2 and He exhausted to the ISS. When one bed is filled, flow is switched to a compan-

ion bed, and the bed full of impurities is regenerated. As reported by Willms [7] and Willms et al. [8], this system has worked reliably in a number of experimental campaigns. During 3-weeks of operation, no ISS plugging occurred due to excessive impurities in the molecular sieve exhaust.

An advantage of this technique is its high reliability. The only moving parts are the solenoid valves supplying liquid nitrogen to the molecular sieve bed. Even if those fail, large thermal gradients ensure that the problem can be dealt with deliberately rather than hurriedly. Furthermore, this is a very forgiving, easily controlled system. It is unaffected by large changes in pressure, flowrate and composition of the feed gas. Note further, that the tritium recovery processing is decoupled from the purification processing. This means that tritium recovery can be performed somewhat leisurely and any problems can be dealt with while still maintaining the high availability needed for the main purification path.

The primary disadvantage of this system is the relatively large tritium inventory adsorbed on molecular sieve while it is in operation. When a bed is first put on-line it adsorbs Q_2 which is later displaced as tritium-containing impurities are adsorbed. In either condition significant tritium is held up on the bed. This fact drives the design toward smaller beds and, thus, more frequent regenerations. Though much smaller than the operational tritium inventory, there will also be a semi-permanent amount of tritium bound up in water which is practically unregenerable from the sieve. If needed, this tritium may be recovered through a series of H_2 exchanges.

Cryogenic molecular sieve beds can accommodate relatively large as well as small total flowrates. However, because impurities accumulate on the beds, only small to moderate impurities flowrates can be handled. High impurities flowrates would result in excessively frequent regenerations.

4.2. Palladium permeators

Palladium-silver alloys have the unique property of having a permeability exclusively for hydrogen isotopes. Rare earth elements may be added to the alloy to further increase the Q_2 permeability and improve other membrane characteristics. When fed a mixture of Q_2 and impurities, such a permeator will produce an impurities-free permeate stream which is pumped by a large vacuum pump to the ISS. The retentate contains all of the impurities (including He) and some Q_2 . This stream is sent to one of the tritium recovery schemes.

The main advantage of permeators is their very low tritium inventory. The only inventory is the small amount of tritium dissolved in the Pd alloy and that in the permeator contained volume (there may, however, be significant tritium heldup in buffer volumes which may be required due to the pressure sensitive nature of permeators). A permeator can also readily accommodate relatively large impurities concentrations. Whereas large impurities flowrates would necessitate excessively large cryogenic molecular sieve beds (to maintain reasonable regeneration cycle times), this would have a minimal effect on a permeator design. The ability of permeators to separate He from Q_2 is also considered an advantage.

Permeators are expected to be less reliable than cryogenic molecular sieve, first, because they are more active in their fuel processing. Secondly, they are continuously linked to their tritium recovery train and, thus, both purification and tritium recovery must operate concurrently with high reliability and availability in order to achieve high reliability and availability of the overall FCU. This requirement could be relaxed by including buffer tanks, but this would offset this technique's biggest advantage – its low tritium inventory. Also permeators are expected to be more difficult to control and more susceptible to process upsets in other parts of the fuel processing system.

Small to moderate total flowrates coupled with moderate to high impurities flowrates and handled well by permeators. Attempting to handle small impurities flowrate in a high total flowrate with a permeator would result in excessive Q_2 in the retentate and a heavy demand on the tritium recovery system.

Carlson et al. [9] indicate that good results have been obtained with a small scale permeator which was successfully operated with various amounts of tritium and impurities for 450 days. Though some mechanical degradation of the palladium tubes was observed, this was not significant enough to affect the permeator's performance. A larger, $\frac{1}{5}$ th ITER-scale palladium permeator has been installed at TSTA and will soon be tested with tritium in the TSTA fuel cycle processing loop.

A general comment is needed here regarding the use of large, efficient vacuum pumps for this and many of the operations discussed below. It has been widely assumed that a scroll pump can provide this function and will pump Q_2 at a rate comparable to its helium pump curves which have been measured. This, however, has not been established by experiment and scroll pumps are now believed to evacuate Q_2 less efficiently than helium. Should this be true, more or larger pumps

will be required to provide a given requirement, resulting in bulkier, more costly systems with reduced reliability and possibly higher tritium inventories in the pump system.

3. Tritium recovery schemes – water intermediate

Cryocondensation, cryogenic molecular sieve and palladium permeators will produce a stream of mostly impurities mixed with some Q_2 . Methods for recovering the tritium from the impurities fall into two categories – those that use water as an intermediate processing step and those that accomplish the recovery directly without a water intermediate.

The first step for all water intermediate methods is to oxidize the stream of impurities and Q_2 . This is accomplished by mixing the impurities with oxygen, converting all Q (diatomic and molecular) to Q_2O in a catalytic reactor, collecting the water on a freezer (or molecular sieve), and exhausting the remaining tritium-free components (CO_2 , N_2 , etc.) to the tritium waste treatment system (TWT). In a final step the freezer is regenerated by heating and sending the steam to one of the units described below for the final recovery of tritium which is readmitted to the main purification path.

This formation and separation of water from the other impurities has been successfully demonstrated by Willms et al. [8] at TSTA. Conversion of all Q_2 into the water form, collection of the water as ice and exhausting of the tritium-free impurities could be performed with no detectable loss of tritium to the TWT.

Recovery of Q_2 from water may be accomplished using hot metal beds, electrolysis, a shift catalyst/permeator combination or a palladium membrane reactor.

3.1. Hot metal beds

Hot uranium (750 K) has been used to recover tritium from water by forming solid uranium oxide and leaving Q_2 in the gas phase. An analogous, yet superior, technique is to use hot magnesium (750 K) to collect the oxygen as magnesium oxide and again leave the Q_2 in the gas phase. This latter method avoids the problems associated with uranium such as its radioactivity, toxicity and pyrophoricity. However, both methods suffer with the problem of producing unregenerable waste and would not be recommended for the large processing requirements associated with machines such as ITER. This may appropriately be used for small processing applications.

During $\frac{1}{5}$ th ITER-scale tests at TSTA, very poor reduction of water over hot uranium was observed. This is believed to be due to formation of an oxide layer at the surface of the uranium powder which greatly impedes the reduction reaction. However, this behavior was not observed by Ache et al. [4] who have successfully reduced water using this technique. This may indicate that there was some other problem with the TSTA beds. Initial TSTA tests using magnesium have been quite successful.

3.2. Electrolysis

Both gas phase and liquid phase electrolysis designs have been proposed. In either case an electrical potential is used to split water into a stream of oxygen and Q_2 . The great advantage of this over hot metal beds is that no waste is produced. The concerns related to electrolysis include scale-up, combustion problems associated with the pure O_2 formed, large tritium inventories for traditional wet cells, and fragility of large ceramic components for high temperature gas phase cells. Carlson et al. [9] report that a small scale gas phase ceramic electrolysis cell has been operated successfully with tritiated water for over a year. A larger, $\frac{1}{5}$ th ITER-scale ceramic electrolysis cell has been installed at TSTA and will soon be tested with tritium in the TSTA fuel cycle processing loop. Work on liquid phase electrolysis has been reported by Bruggeman et al. [28] and Giroux [29].

3.3. Shift reaction / permeation

The water–gas shift reaction, $CO + H_2O \rightleftharpoons CO_2 + H_2$, can be used to recover H_2 from water. This reaction only proceeds to partial conversion so, at equilibrium, some H_2O is still present. By removing the H_2 from the reacting system with, for instance, a palladium permeator the equilibrium can be upset and further shift can be accomplished. This sequence of shift followed by H_2 removal can be repeated until the tritium has been sufficiently removed from the system. Such a system has been examined and proposed by Penzhorn et al. [10], Glugla et al. [11,12] and Yoshida et al. [13].

3.4. Palladium membrane reactor

An extension of the shift/permeation concept is the use of a membrane reactor. Membrane reactors have

been investigated since the late 1960s for applications in areas other than fusion technology. They typically consist of a plug-flow catalytic reactor with walls composed of a membrane material. This is useful for shift reactions which only go to partial completion since the equilibrium can be upset continuously down the length of the reactor (rather than stage-wise as in the shift/permeation case). This technique has been applied successfully in other fields, for example, to hydrogenation and dehydrogenation organic reactions (see Gryaznov [14], Ito et al. [15], Ito [16], and Ito and Govind [17]. Theoretical treatments of this operation have also been presented by Mohan and Govind [18,19,20].

For tritium recovery, a palladium-walled reactor is filled with water-gas shift catalyst. Though more favorable equilibrium is achieved at lower temperatures, both the permeator and certain shift catalysts work reasonably well in the 573 to 673 K range. CO is used to sweep steam to the membrane reactor. Therein, the water-gas shift reaction may proceed to completion because the Q_2 is being continually pumped away by a vacuum pump and returned to the main purification path.

The primary problem unique to this design is that the palladium membrane reactor has not been demonstrated in tritium service. However, having already been successfully tested in analogous situations, it is expected that the membrane reactor will work in tritium service. Also of concern is the addition of more carbon to the system. It may be that sufficient ^{14}C is generated in the fusion reactor so that all carbon will need to be collected. Thus, the addition of more carbon (as CO) will place an increased load on the carbon collection system.

4. Tritium recovery schemes – direct tritium recovery

There are a number of reasons to avoid the production of water in the tritium recovery train. These include increased biological hazard, condensation, undesirable reactivity (e.g. the formation of nitric acid), possible corrosion problems, and increased operational complication and tritium inventory because of the added step of water formation and collection. It is clear that, in the absence of other overriding considerations, it is preferable to avoid water production. The following techniques – shift catalyst/permeator combination, palladium membrane reactors and the HITEX process – include no water formation. It must be noted that experiments with the following methods are only

now beginning. Recovery efficiency with them remains to be demonstrated.

4.1. Shift catalyst / permeator

This scheme employs a similar, but more ambitious use of the shift catalyst/permeator concept presented above. In the previous case only one reaction, the water-gas shift reaction, was being utilized because all Q had been converted to the water form. In this case direct conversion of the impurities is accomplished via a number of additional reactions such as the steam reforming reaction ($\text{CH}_4 + \text{H}_2\text{O} \rightleftharpoons \text{CO} + 3\text{H}_2$), the methane cracking reaction ($\text{CH}_4 \rightleftharpoons \text{C} + 2\text{H}_2$), and the reverse ammonia synthesis reaction ($2\text{NH}_3 \rightleftharpoons \text{N}_2 + 3\text{H}_2$). These individual reactions have been studied for decades and, recently, study has begun on these reactions in the context of tritium systems. Xu and Froment [21,22] describe the type of treatment necessary for analyzing this complex reaction system.

In this design, impurities are sent from the purification system directly to the shift catalyst bed or beds using CO and/or He as a carrier gas. All of the reactions shift from Q_2 -containing molecules such as CQ_4 , $Q_2\text{O}$ and NQ_3 toward CO_x , N_2 (maybe NO_x) and Q_2 . Removing the Q_2 with a palladium permeator enables further shift to occur. Stage-wise shift followed by permeation is repeated until sufficient tritium recovery is attained. In this manner all hydrogen isotopes can be recovered without going through the intermediate step of forming water.

While this is conceptually quite feasible, there are many unknowns. For instance, coking may be a problem and it is unclear what will happen when compounds other than the simple model compounds listed above are encountered. As explained earlier, ^{14}C is also a concern.

Besides avoiding water formation, this system also accomplishes tritium recovery in a single step, is easier to operate and reduces the time averaged tritium inventory. Penzhorn et al. [30] report that an ITER-relevant scale experiment based on these principles is beginning.

4.2. Palladium membrane reactor

It is also possible to accomplish the direct recovery of tritium from impurities with a palladium membrane reactor packed with shift catalyst. As explained earlier, this would enable the shift reaction equilibria to be upset continuously as the Q_2 is removed from the reacting environment. This would enable tritium recovery

ery to be accomplished in a manner conceptually similar to the shift catalyst/permeator option, but in a more compact, easier to operate process.

While many features make this an attractive tritium recovery option, there are a number of concerns beyond those expressed for the shift catalyst/permeator. A catalyst that might perform all of these reactions at once (e.g. a mixture of water-gas shift and steam reforming catalyst) has not been tested. Also, since the reactor is contained with the permeator, the catalyst or catalyst blend would have to work at the palladium permeator temperature, 573 to 673 K.

4.3. HITEX

Another tritium recovery scheme which avoids water formation is the "HITEX" (HI Temperature EX-change) process which has been proposed by Busigin et al. [23]. The tritium-containing impurities are sent to a hydrogen-rich processing loop which includes a high temperature isotopic exchange catalyst. Therein, reactions such as, $\text{CH}_3\text{T} + \text{H}_2 \rightleftharpoons \text{CH}_4 + \text{HT}$ occur. Following this catalyst is a permeator which separates Q_2 from the impurities. The pure Q_2 , containing mostly H and a little D and T, is sent to the hydrogen column in the ISS. For designs such as ITER, this additional H_2 may have a minimal impact on the large ISS hydrogen column. When a batch of gas has been sufficiently stripped of tritium, the remaining impurities and hydrogen are vented to the TWT.

This technique has advantages similar to the palladium membrane reactor described above. Furthermore, it does not add carbon to the system and will, therefore, not tax a carbon collection system.

The disadvantage of this system is its use of large amounts of hydrogen. This necessitates the use of a fairly large permeator-pump combination and results in an increased load on the ISS. For fuel processing systems which use a relatively small ISS, this may not be a viable option. To reduce the size of the permeator-pump combination, relatively high pressure may be used. This high pressure in a tritium system is a disadvantage.

5. Process selection

Determining which process to use for a given application depends on a number of factors including the:

- overall scale of the operation,
- operating scenarios,
- amount of waste that is acceptable,

- degree of reliability required,
- degree of proveness required,
- impurities compositions and flowrates,
- Q_2 flowrates,
- necessity of separating He in FCU,
- size of ISS,
- willingness to accept water as a process intermediate.

5.1. Purification

The first decision which must be made is the choice or choices for purification. For a system such as ITER there will be multiple unique streams which require cleanup. These include gases from the torus vacuum system, the neutral beam system, and the pellet injector system. The first stream is characterized by a relatively low overall flowrate, but a relatively high impurities concentration. The other two streams have relatively high overall flowrates, but minimal impurities concentrations. The neutral beam stream is predominantly D_2 while the pellet injector gas is mostly H_2 . In current design work both palladium permeators and cryogenic molecular sieve are being considered for purifying the first stream, and the other two streams will most likely be purified using separate cryogenic molecular sieve bed trains.

The designer must choose purification schemes which are appropriate to each of the streams encountered. Where possible streams may be combined to consolidate purification operations. In some cases, where reactor operating scenarios will vary significantly, more than one purification scheme may be needed for a single stream.

5.2. Tritium recovery

With the purification stream chosen and sized, the tritium recovery system must be considered. It is likely that only one tritium recovery system will be needed since it will be able to serve a number of purification scenarios. The tritium recovery system may also serve the needs of purification operations located outside the FCU such as in a blanket tritium recovery system.

6. Conclusions

There are a number of ways to perform the functions of an FCU – purification and tritium recovery. FCU designers may choose and configure these methods in a manner which best meets the needs of a given

application. All four of the purification methods presented have ranges of applicability in which they may be found useful. For tritium recovery the methods which avoid water as an intermediate are generally considered more desirable but so far suffer from being more experimental in nature. Refinements in all of these techniques are expected as research continues. Furthermore, new methods are likely to be proposed. Robust, well-tailored FCU's can be realised using these various processing options.

7. Summary

This paper has identified the two main functions of a fuel cleanup system – purification and tritium recovery. Purification may be accomplished by cryocondensation, hot metal beds, cryogenic molecular sieve and/or palladium permeators. Tritium recovery may be performed either with or without a water intermediate. Methods which required a water intermediate include hot metal beds, electrolysis, shift catalyst permeator and palladium membrane reactors. Preferable, but more experimental methods of tritium recovery which avoid a water intermediate are shift catalyst/permeators, palladium membrane reactors, and HI-TEX.

Each of these methods have been described and their strong and weak points identified. Considerations related to choosing the right combination of methods for designing a complete FCU have been presented.

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